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Scope of Research

Our research interest is to understand optical and quantum properties of nanometer-structured materials and to establish opto-nanoscience for creation of innovative functional materials. Optical properties of semiconductor quantum nanostructures and strongly-correlated electron systems in low-dimensional materials are studied by means of space- and time-resolved laser spectroscopy. The main subjects are as follows: (1) Investigation of optical properties of single nanostructures through the development of high-resolution optical microscope, (2) Development of nanoparticle assemblies with new optical functionalities, and (3) Ultrafast optical spectroscopy of excited states of semiconductor nanostructures.

KEYWORDS

Femtosecond Laser Spectroscopy
Carbon Nanotubes
Semiconductor Nanoparticles

Transition Metal Oxides
Semiconductor Nanostructures



Selected Publications

Matsunaga, R.; Matsuda, K.; Kanemitsu, Y., Observation of Charged Excitons in Hole-doped Carbon Nanotubes Using Photoluminescence and Absorption Spectroscopy, *Phys. Rev. Lett.*, **106**, [037404-1]-[037404-4] (2011).

Yamada, Y.; Yasuda, H.; Tayagaki, T.; Kanemitsu, Y., Temperature Dependence of Photoluminescence Spectra of Undoped and Electron-doped SrTiO₃: Crossover from Auger Recombination to Single-carrier Trapping, *Phys. Rev. Lett.*, **102**, [247401-1]-[247401-4] (2009).

Matsunaga, R.; Matsuda, K.; Kanemitsu, Y., Evidence for Dark Excitons in a Single Carbon Nanotube Due to the Aharonov-Bohm Effect, *Phys. Rev. Lett.*, **101**, [147404-1]-[147404-4] (2008).

Hosoki, K.; Tayagaki, T.; Yamamoto, S.; Matsuda, K.; Kanemitsu, Y., Direct and Stepwise Energy Transfer from Excitons to Plasmons in Close-packed Metal and Semiconductor Nanoparticle Monolayer Films, *Phys. Rev. Lett.*, **100**, [207404-1]-[207404-4] (2008).

Observation of Charged Excitons in Hole-doped Carbon Nanotubes

Carbon nanotubes are one of the excellent materials for studying the optical properties of excitons, because of their unique band structures and large exciton binding energies. We report the first observation of trions (charged excitons), three-particle bound states consisting of one electron and two holes, in hole-doped carbon nanotubes at room temperature. When p-type dopants are added to carbon nanotube solutions, the photoluminescence and absorption peaks of the trions appear far below the E_{11} bright exciton peak, regardless of the dopant species. The unexpectedly large energy separation between the bright excitons and the trions is attributed to the strong electron-hole exchange interaction in carbon nanotubes.

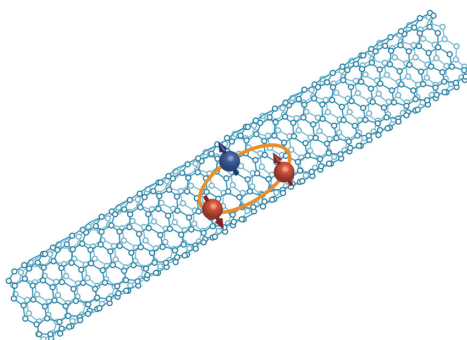


Figure 1. Schematic illustration of charged excitons in carbon nanotubes.

Quantized Auger Recombination of Biexcitons in CdSe Nanorods

Fabrication and characterization of semiconductor nanocrystals (NCs) have been extensively studied due to interest both in the fundamental physics and potential applications in optoelectronic devices. We studied the recombination dynamics of biexcitons in elongated CdSe nanocrystals (nanorods) using time-resolved photoluminescence (PL) and transient absorption (TA) spectroscopy. The decay times of the PL and TA signals decrease with increasing nanorod length. Under weak excitation, the PL decay is faster than the TA decay, and the nonradiative hole trapping determines the PL decay in several hundreds of picoseconds. Under intense excitation, the PL decay curves are similar to the TA decay curves, and the rapid biexciton decay is caused by nonradiative Auger recombination. A clear correlation is observed between the Auger recombination coefficient of the biexcitons and the average PL lifetime of the single excitons. Moreover, the Auger recombination lifetimes of the biexcitons are shorter in nanorods than in spherical nanocrystals of the same volume.

Our study clarified that the Auger recombination rate is strongly affected by a high surface-state density.

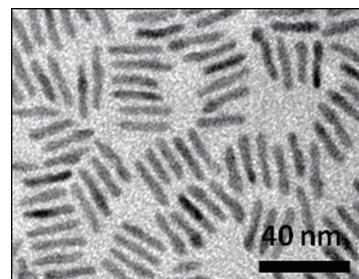


Figure 2. Transmission electron microscope image of elongated CdSe nanocrystals (nanorods).

High-Density Carrier Dynamics in Ge/Si Quantum Dots

Quantum dots (QDs) have attracted attention because of their interesting physical properties and potential applications in optoelectronic devices such as light emitters and solar cells. In QDs, physical processes of generation, relaxation, and recombination of carriers are determined by their nanostructures and differ from those in bulk crystals. We studied photoluminescence (PL) dynamics in thermally annealed Ge QDs in silicon crystals to determine the role of Ge-Si intermixing in carrier recombination processes. PL decay measurements revealed that the fast-decay component appeared under high-density photoexcitation, indicating nonradiative Auger recombination. By varying the annealing temperature of the QDs, the fast Auger decay time became larger than that of the as-grown QDs. This finding indicates that the Auger recombination rate is affected by Ge-Si intermixing due to thermal annealing.

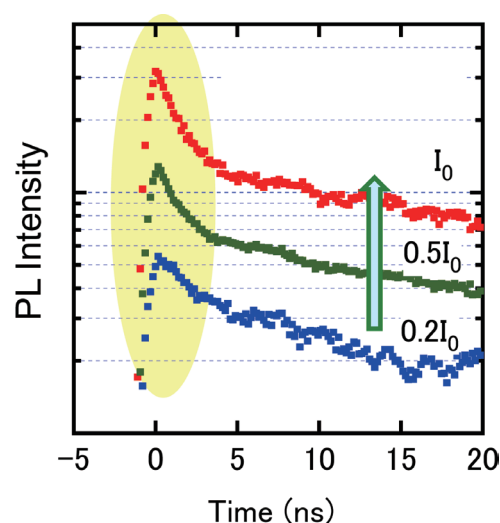


Figure 3. PL decay profiles at different photoexcitation intensities.